

REMARKS

The Examiner has again rejected claims 1-3 under 35 U.S.C. 103 for obviousness over Johnson et al (U.S. 5,520,708), in view of Kovacs, et al. Applicants respectfully urge that this is not the case, and it is submitted that this ground of rejection should be withdrawn in view of the attached amendments and arguments.

The presently claimed invention, as amended above, provides a method for improving the long term stability of biodiesel, comprising:

- (a) forming a reaction mixture comprising a crude methyl ester, by transesterification of a vegetable or animal fat or oil with methanol,
- (b) forming a layer containing the crude methyl ester of step (a), and separating the layer from the rest of the reaction mixture,
- (c) intensively inline mixing the crude methyl ester layer obtained in step (b) at temperatures between 25 and 60°C with a strong acid or with a mixture of a strong acid and a complex former, to form an emulsion, and
- (d) separating an ester layer from the emulsion formed in step (c), and then subjecting the separated ester layer to a thorough water wash and a subsequent drying.

Applicants submit that no new matter is entered. Support for step (a) wherein a reaction mixture comprising crude methyl ester is formed by transesterification of a vegetable or animal fat or oil with methanol, can be found in the specification at page 3 in the first paragraph. Support for step (b) and the separation of a crude methyl ester layer from the reaction mixture can be found in the specification at page 4, in the fourth full paragraph which states that the crude methyl ester formed by transesterification is initially present in a mixture with split-off glycerin. This paragraph further states that that crude methyl ester is separated from the heavy phase containing the glycerin and residual catalyst. In the fifth paragraph on page 4, it is stated that the crude ester phase is fed into an intensive mixing apparatus. Support for step (c), including the intensive inline mixing of the crude methyl ester layer at temperatures between 25 and 60°C with a strong acid or mixture of a

strong acid and a complex former can be found in the specification at page 3, in the second full paragraph. Further support for a strong acid or mixture of a strong acid and a complex former is shown in the last paragraph of page 4, which continues to page 5. Support for the separating of an ester layer in step (d) from the formed emulsion can be found in the specification at page 3, first paragraph, which states that "the ester layer separated from the emulsion layer", and "is subjected to a thorough water washing and is subsequently dried". Additional support can be found on page 5, in the first and second full paragraphs which discuss ester layer formation and water washing. It should be noted that claim 2 has been amended for clarity, to correct the typographical spelling of ptoluenesulfonic to now read "p-toluenesulfonic", and to correct the shorthand term EDTA to now read "ethylenediaminetetraacetic acid" as is well known in the art.

Regarding the present rejection under 35 U.S.C. 103, Applicants strongly urge that the presently claimed invention is not taught or suggested by the cited art. The Examiner first cites Johnson, et al. the reasons stated in the office action. First, it is again submitted that Johnson relates to a completely different subject matter than the present application. Johnson provides a method for reducing the crystallization temperature of a fatty acid oil ester-petroleum distillate fuel blend, while the present invention serves to improve the long-term stability of biodiesel. Furthermore, Johnson combines petroleum distillates, such as a diesel fuel and other middle petroleum distillates, with esters of branched chain alcohols such as isopropyl esters, 2-butyl esters, and/or tertiary butyl esters synthesized from fats and oils containing fatty acid triglycerides. While Johnson may use methanol as an exchange alcohol, and strong acids such as sulfuric acid and hydrogen chloride, it is submitted that the purpose of using these reagents is very different from the present invention and provides a different result. Specifically, the transesterification step (a) of the present claim 1 is well known in the art in and of itself. However, it is asserted that the combination of steps (a)-(d) has *not* been heretofore known in the art, particularly for improving the long term stability of a biodiesel. These steps include: separating a crude methyl ester layer from the reaction mixture of step (a); intensively inline mixing the crude methyl ester layer at temperatures between 25 and 60°C with a strong acid or with a mixture of a strong acid and a complex former, to form an emulsion; and separating an

ester layer from said emulsion and subjecting it to a water wash and drying step. This sequence of process steps is not taught or suggested by Johnson, and it is therefore non-obvious in view of the cited art.

The Examiner asserts that there is no reason to believe that the process of Johnson would not also improve the stability of a biodiesels fuel. Applicants urge that this is not the case. That is, even if one presumes that Johnson would unintentionally and unrecognizably produce an effect in their reaction mixture equivalent to the present application, the effectiveness of this would be negligible compared to the present application. The reason for the increased *stability* of the biodiesels produced according to the present application is the elimination of crystallization nuclei by the action of the acid. This is achieved by a more intense washing and, most importantly, a hydrophilization of the crystallization nuclei. Having recognized this mechanism for the flocculation in biodiesels and the possible counter action, the present application presents a process which is optimally suited for this method. Johnson only has a single distribution between phases and a usual amount of acid catalyst. This will result in a biodiesel product prone to flocculation *like all the biodiesels of the state of the art*. The present application features a second distribution equilibrium and a much higher acid content therein. The latter aids in the hydrophilization of the crystallization nuclei and the former improves extraction of the hydrophilized nuclei. According to the Nernst distribution law, the extraction effect of two steps instead of one, and an increased concentration difference between the phases will result in a much better extraction.

An important difference exists between the use of strong acid in the Johnson disclosure as compared to the present invention. Johnson teaches the use of strong acids such as sulfuric acid and hydrogen chloride as *acid catalysts*, as also pointed out by the examiner. However, in the present invention, the strong acids are employed to aid splitting the undesired soaps, which are considered as one kind of "crystallization nuclei" contributing to the long term instability of biodiesel. Therefore the presence of the acids in Johnson and the present invention relate to completely different mechanisms. Furthermore, in Johnson all the steps of acid application, water washing, stirring, and emulsion formation,

occur during a transesterification phase, which is before the downstream workup and purification process described in the current invention to improve the long term stability of biodiesels.

The Examiner asserts that in the reaction scheme of Johnson, the same effect as the present invention is achieved because their acid catalyst is still present when the ester is mixed. Applicants respectfully urge that this is not the case. First, Johnson does not mix their ester while the original amount of acid is still present. Example 1 clearly teaches that the oil and the alcohol are vigorously stirred together with sodium alcoholate. This is done to improve the reaction speed, because the oil and alcohol don't mix well. Johnson monitors the reaction by TLC and adds, after completion of the esterification, the hexane/water mix in order to induce phase separation. Thereafter the ester phase is washed with water. There is no mention of a further vigorous stirring of the reaction mixture after the addition of the hexane/water mix or during the washing. The main part of the acid, which according to Johnson is present only in amounts of 0.1 to 0.5 wt.% based on the weight of the oil, is discarded with the separated lower aqueous phase. Thus, there is a major difference in the emulsion formed by the present application and the droplets dispersed the vigorous stirring Johnson describes. The present application forms an emulsion of acid in the separated ester rich phase. In this phase there are only traces of the alcohol and by-products left, and it is emulgated with pure (or diluted) acid. In contrast to this, Johnson has a completely different composition, no matter at which time the Examiner assumes the mixture to be of equal quality. Should it be the reaction mixture before the addition of the hexane/water mix, then there are still all by-products and the excess alcohol present. Should it be the washing mixture, then there are only traces of the acid left, because the acid is separated with the lower aqueous phase. The method of the present invention is therefore completely different to the method of Johnson and results in completely different emulsion mixtures. Therefore the reactions taking place and the distribution behavior are completely different as well.

An important feature of the present invention is to intentionally form an emulsion by the use of an intensive inline mixing apparatus. As the examiner appreciates, the

micelles created by an emulsion ensures a very large surface area between the ester phase and the aqueous phase, and thus an enhanced mixing effect and the removal of the impurities is achieved. The emulsion then breaks and a stable interphase between a washed ester phase and the wash phase is formed. As stated in Applicant's previous paper, the transesterification step itself is of no particular interest to the present invention, but rather combination of step (a) with the downstream purification steps (b)-(d) are the focus. It is submitted that it is not obvious for those skilled in the art to use acid and emulsion formation to remove the impurities in the crude biodiesel based on the teaching of the cited prior art documents, since all of these references focus only on the transesterification of certain oils with methanol. The presence of crystallization nuclei and the problem of long term instability have not even been mentioned therein, let alone this would require a fundamentally different knowledge of the properties and usage of acids.

The Examiner asserts on pages 4-5 of the office action that the steps of the present invention appear to be performed by Johnson in their Example 1 at column 4, lines 19-31. Applicants respectfully disagree. As stated above, the downstream purification steps are key features of the present invention. However, again, Johnson does not teach steps (b)-(d) of the presently amended claims which include: separating a crude methyl ester layer from the reaction mixture of step (a); intensively inline mixing the crude methyl ester layer at temperatures between 25 and 60°C with a strong acid or with a mixture of a strong acid and a complex former, to form an emulsion; and separating an ester layer from said emulsion and subjecting it to a water wash and drying step. That is, the present claims require two distinct emulsion separation steps, while Johnson teaches a *single* separation step in their Example 1.

Additional strong evidence that the effect taking place in the present application and the effect asserted by the Examiner to be inherently achieved by Johnson cannot be the same, is the fact that Johnson focuses on a method to improve the low temperature stability of diesel fuels by mixing them with vegetable oil esters which are produced by methods well known in the state of the art. The applicants have however described that all of the

biodiesels known in the state of the art suffer from deficient long-term stability due to flocculation. If Johnson had been able to produce a diesel product without this disadvantage he would undoubtedly have mentioned it as this would solve a major problem of today's biodiesels.

Even if an effect of a reduction of crystallization nuclei by the action of the acid had arguably been achieved by Johnson, the present invention would still not have been obvious. The crucial understanding is that the flocculation is caused by crystallization nuclei and the achievement of the invention is a method to eliminate them. The method may arguably make use of an effect which might be present in the methods of the state of the art, but only by applying the method claimed by the applicants does the effect produce a useful and detectable result.

As the Examiner agrees on page 5 of the office action, Johnson makes no mention of forming an emulsion. In an effort to fill this void of Johnson, the examiner cites Kovacs, et al (WO 03/040081), which allegedly comments on the methods developed by Johnson. The approach disregards the original Johnson disclosure and relies on an indirect resource. The scope of protection of a patent is defined by its claims, which are supported and/or interpreted by its description and drawings. To allow the interpretations extends beyond the original disclosure and by not even a well established teaching of common knowledge leads undoubtedly to great uncertainty.

The second paragraph of page 3 by Kovacs as cited by the examiner, actually refers to two documents, namely U.S. patents No. 5,520,708 and 6,015,444. So, the comments stated in the paragraph do not exclusively refer to U.S. patent 5,520,708 by Johnson et al. Surprisingly, U.S. 6,015,444, does not pertain to the production of biological oils, rather it belongs to a totally unrelated area of mechanical engineering, namely "An apparatus and system for venting a transmission". The foregoing leads to doubt and that there might very well have been a mistake in referencing the background state of the art in the text by Kovacs.

Furthermore, even the presence of an emulsion as taught by Kovacs was hypothetically combined with Johnson as stated by the Examiner, an emulsion supposedly formed by such a combination would still be conducted only during the transesterification stage and serves only to mix the reactants including catalysts intensively. It has literally been stated that "The lifespan of the emulsion formed in the homogenizer enables transesterification to proceed to equilibrium conversion before the emulsion segregates in the settler." Completely differently, the present invention utilizes an emulsion after transesterification to remove impurities. Therefore the addition of Kovacs with Johnson can by no means negate the non-obviousness of the present invention. Additionally, as stated above, a key feature of the present invention relates to the steps of: separating a crude methyl ester layer from the reaction mixture of step (a); intensively inline mixing the crude methyl ester layer at temperatures between 25 and 60°C with a strong acid or with a mixture of a strong acid and a complex former, to form an emulsion; and separating an ester layer from said emulsion and subjecting it to a water wash and drying step. These steps (b)-(d) of the presently amended claims are absent from both Johnson and Kovacs, and thus the present invention fails to be obviated by the cited art. In addition, these references address completely different technical problems, which then lead to totally different methods utilized to solve their problems. It is thus urged the present invention is not obvious in view of Johnson and Kovacs.

The examiner is further of the view that both the Johnson and the present invention teach washing the reaction mixture and subject the mixture to drying. Once again, this position is inappropriate since these steps are involved in different stages in the two inventions, with one being after the transesterification stage and the other one being after the removing the impurities which cause the long term instability of biodiesel.

Regarding the Examiner's comments on claim 3, applicants wish to respectfully point out that a separatory funnel is neither a wash column nor does it work with the counter current principle. A separatory funnel is *by definition* a single extraction stage with works bath-wise. The two immiscible liquids are introduced into the funnel, intensively mixed, and then allowed to separate again. There is no current at all.

For all of the above reasons, it is respectfully urged that the present claims are non-obvious in view of the cited art, and the 35 U.S.C. 103 rejection should be withdrawn.

The undersigned respectfully requests re-examination of this application and believes it is now in condition for allowance. Such action is requested. If the Examiner believes there is any matter which prevents allowance of the present application, it is requested that the undersigned be contacted to arrange for an interview which may expedite prosecution.

Respectfully submitted,



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I hereby certify that this paper is being facsimile transmitted to the Patent and Trademark Office (FAX No. 571-273-8300) on October 6, 2009.



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